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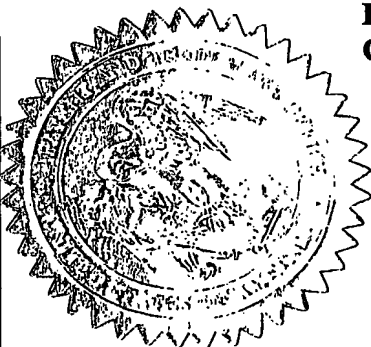
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11062 U.S. PTO

Provisional Application Cover Sheet

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This is a request for filing a PROVISIONAL APPLICATION under 37 C.F.R. § 1.53(b)(2).

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Inventor(s)/Applicant(s)				
Last Name	First Name	Middle Initial	Residence (City and either State or Foreign Country)	
Yodh Islam	Arjun Mohammad	G F	Merion, PA Philadelphia, PA	
Title of the Invention (280 Characters Maximum)				
Alignment of Single-wall Carbon Nanotubes: Nematic Nanotube Gels				
Correspondence Address				
University of Pennsylvania Center For Technology Transfer 3160 Chestnut Street Suite 200				
City: Philadelphia	State: Pennsylvania	Zip Code: 19104 - 6283	Country: US	
Enclosed Application Parts (check all that apply)				
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The invention was made by an agency of the United States Government or under a contract with an agency of the United States Government.

☐ No

☒ Yes, the name of the U.S. Government agency and the Government contract number are:

NSF Grant Number DMR0079909

Respectfully submitted,

Signature: M. F. Islam
Typed or Printed Name: Mohammad Islam

Date: 10/18/02

☐ Additional inventors are being named on separately numbered sheets attached hereto.

PROVISIONAL APPLICATION FILING ONLY

1:879 U.S. PTO

60/419882

Single-wall carbon nanotubes (SWNTs) possess highly anisotropic and unique mechanical, electrical and thermal properties [2–14]. Applications for nanotubes utilizing these attributes generally fall into two categories: those requiring isolated tubes [5, 9–11], and those requiring ensembles of tubes [12, 13, 15–22]. In the latter case, specially for composite materials, a high degree of tube alignment are desired [23]. Aligning tubes is difficult however. With few exceptions [17, 22], the vast majority of solution- and solid-phase mixtures are isotropic [15, 16, 18–21]. Stable nematic-like phases of SWNTs, in which tubes are oriented along a single direction, have been elusive.

I. BACKGROUND AND PREPARATIONS

Our nanotubes were obtained in purified form from Tubes@Rice (laser-oven SWNTs, batch P081600 [24]). The purified sample had > 90 wt% SWNTs. We dispersed SWNTs in water using a low-power high-frequency bath sonicator (12 W, 55 KHz) and surfactant molecules, sodium dodecylbenzene sulfonate (NaDDBS; $C_{12}H_{25}C_6H_4SO_3Na$) [25]. The nanotube-to-surfactant ratio was 1 : 10 by weight. These nanotube dispersions had very high yield of single tubes ($\sim 90 \pm 5\%$) with average length $L_{\text{mean}} \sim 516 \text{ nm} \pm 286 \text{ nm}$ [25]. It is not critical to use laser-oven SWNTs. We have also used a different nanotubes, called HiPCO (Carbon Nanotechnologies Inc., batch 79 [26]; $L_{\text{mean}} \sim 165 \text{ nm} \pm 95 \text{ nm}$ [25]) and obtained similar results.

Most, but not all of our experiments used a gel consisting of polymerized *N*-isopropyl acrylamide monomer (NIPA; 700 mM), *N,N'*-methylenebisacrylamide (cross-linker agent; 8.6 mM), ammonium persulfate (initiator; 3.5 mM) and *N,N,N',N'*-tetramethylethylenediamine (accelerator at 295 K; 0.001% v/v). All ingredients were obtained from PolySciences Inc. (Warrington, Pa) and were used without further purification. To prepare SWNTs-NIPA gels, we first mixed SWNTs dispersions and all gel reagents except the initiator in water. The SWNTs concentrations ranged from 0.04 mg/ml to 0.78 mg/ml. We could not use SWNTs concentrations higher than 0.78 mg/ml in NIPA because the NIPA monomer did not gel. The gel initiator was then added to the mixture which was then vortexed for 15 seconds. The polymerization takes ~ 1 hour. The vortexed pre-gel solutions were loaded into rectangular capillary tubes with inner dimensions (length \times width \times thickness) of $\sim 4 \text{ cm} \times \sim 4 \text{ mm} \times 0.2 \text{ mm}$ and a wall thickness of 0.2 mm. Fig. 1 show

an schematic of gel structure after polymerization in the presence of cross-linker.

To align the nanotubes outside of a magnet, the pre-gel solution were allowed to polymerized at 295 K for ~ 3 hour on top of a lab work-bench. To align the tubes inside of a magnet, the capillary tubes with SWNTs-NIPA gel were placed inside a 9 Tesla magnet and at 295 K for longer than the duration of the polymerization process (~ 2 hrs). The initial gelation process appeared to lock nanotubes into place, producing a dilute tube distribution with random location. The tube orientation was random when the gel polymerized outside of a magnet; the tube orientation was parallel to the applied field when gel polymerized inside a magnet. We suspect the tubes could not diffuse over long distances in the gel, but could reorient and move short distances with relatively small energy cost.

Images depicting birefringence in our nematic nanotube gels were obtained with samples situated between crossed-polarizers in the microscope on a rotating stage. The crossed-polarizer measurement is sensitive to birefringence in the sample, which arises when nanotubes align locally. The aligned regions appear bright in the image; isotropic regions appear dark. For clarity we set the pass axes for the input and output polarizers to be along the x and y directions respectively; the light transmission direction was along z ; the sample was rotated in the xy -plane. By rotating the stage, we obtained information about the direction of alignment of nanotubes. We did not quantitatively measure the degree of nanotube alignment. However, by keeping the microscope bulb intensity and the video gain/offset the same for the full set of images, we obtained semi-quantitative information about the degree of alignment of nanotubes in the NIPA gel from the relative intensity differences between various images or regions within a image. An increase in the degree of nanotube alignment is manifested as an emerging bright domain or an increase in the brightness of a domain. Bright-field images of the nanotube needle or ropes in the nematic nanotube gels were obtained without cross-polarizers. We visualized the birefringence and the structures within the sample using a Leica DMIRB inverted optical microscope with a $10\times$, $5\times$ and $1.6\times$ air objectives. The samples were imaged using a CCD camera (Hitachi, model KP-M1U, 640×480 pixels) and recorded directly into a computer hard-drive using a 8-bit video frame grabber (model CG7, Scion Corporations, Frederick, Maryland).

The magnet used to align the nanotubes was a super-conducting magnet (Quantum Design, San Diego, CA) where the magnetic field could be varied between -10 Tesla to $+10$ Tesla and the temperature could be varied between 40 K to 373 K. To align the nanotubes,

we loaded the capillary inside the magnet immediately after the polymerization of NIPA gel was initiated and was left inside the magnet at 9 Tesla for longer than the duration of polymerization (~ 2 hour).

II. DESCRIPTION OF TECHNOLOGY

I. Method 1: Local alignment of SWNTs by shrinking the SWNTs-NIPA gel
To induce nematic-like structures in SWNTs-NIPA gel, we immersed the capillary tubes with SWNTs-NIPA gel inside glass vials containing 20mM Trizma buffer and placed the entire sample assembly inside an oven at 323 K. The polymer network in the gel became hydrophobic around 323 K. The gel then reduced its volume by expelling water and therefore, the effective volume fraction of the locked SWNTs in the gel increased. For sufficiently large initial nanotube concentrations the tubes aligned locally. We then took out the capillary tube containing shrunk SWNT-NIPA gel, removed the expelled water and imaged the sample under the microscope. Removal of expelled water is important because that prevented the gel from swelling to its pre-shrunk volume as the gel temperature lowered to room temperature (~ 295 K) and the polymer networks became hydrophilic. We refer to this local alignment of SWNTs in NIPA gel as a "quasi-isotropic-nematic" transition.

In Fig. 2 we show a photograph of the SWNTs-NIPA gels and NIPA gels with surfactant alone (7.8 mg/ml NaDDBS) before and after shrinking. Typical pre-shrunk sample dimensions (length \times width \times thickness) were ~ 4 cm \times 4 mm \times 0.2 mm and shrunk dimensions were ~ 2 cm \times 2 mm \times 0.1 mm. The sample in Fig. 2(a) had a high initial nanotube concentration (0.78 mg/ml) and the material undergoes the quasi-isotropic-nematic transition immediately after shrinking. The sample in Fig. 2(b), by contrast, initially contained a dilute mixture of nanotubes (0.23 mg/ml) and the quasi-isotropic-nematic transition was not observed immediately after shrinkage. It is evident from the photograph that the volume change before/after shrinking is large ($\sim 8\times$); the shrunken gels in Fig 2(a) and 2(b) also appear darker, because the tube concentration is higher and the tubes absorb visible light.

In Fig. 3 we show one of our concentrated samples as a function of angle. All the images were taken with fixed microscope bulb intensity and video gain and offset. This sample had an initial tube concentration of 0.78 mg/ml, and was allowed to sit for 4 days after shrinking. The gel exhibited a maximum birefringence when its edge was oriented

45 degrees with respect to the input polarizer pass axis. Liquid crystal like defects were observed near the edges with the sample; clearly visible when the sample was in vertical (0 degree) or horizontal (90 degrees) orientations. Clearly there was greater tube alignment near the gel edges. Evidently the director tends to align near the walls, perhaps as a result of boundary effects. The darker regions in the center of the sample could indicate tube disorder or tube alignment in the z-direction. To distinguish these possibilities we rotated the sample between 10 to 60 degrees about the y-axis. We did not however, observe significant changes in the central birefringence profile. Most likely, the central dark regions were disordered.

The essential features of our concentration- and time-dependent observations are summarized in Fig. 4. Again, we kept the bulb intensity and video gain/offset the same as before. All of the samples were isotropic before shrinking; light transmission was zero. We only observed birefringence in samples that shrank. Twenty minutes after shrinking only the highest initial concentration (0.78 mg/ml) sample exhibited birefringence. As time passed though, the samples slowly evolve. Alignment clearly started at the edges of the sample and migrated inward. After one day the sample critical concentration for birefringence near the edge was approximately 0.54 mg/ml. After 2 days the critical concentration for birefringence had dropped further. The degree of nematic alignment in the samples was found to increase and the critical concentration for nematic alignment decrease, respectively with increasing time after shrinking the gel.

We should point out that these quasi "isotropic-nematic" transitions differed from lyotropic transitions of suspended hard rods [27] in some respects. The transition nanotube volume fractions were lower than expected based on nanotube behavior in water alone, suggesting the gel network plays a significant role in increasing the effective tube interaction and/or local concentration.

So far, we kept the polymerization temperature at 295 K. At this temperature, we expected the gel network and the tube distribution within the gel to be homogeneous [28]. However, if we polymerize the NIPA monomer at a higher temperature (~ 304 K), we expect the nanotubes to micro-phase separate into regions of nanotube rich/gel poor and vice versa. At high enough nanotube concentrations, the nanotubes in nanotube rich/gel poor region can align to become nematic. We have observed such behavior for other rod-like molecules (fd virus) in NIPA gel.

II. Method 2: Nanotube alignment via water extrusion from SWNTs-NIPA gels To extrude water from SWNTs-NIPA gels, we placed the gel-filled capillary tubes inside a vacuum jar and slowly evacuated the jar using a vacuum pump. The experimental setup is shown in Fig. 5. Initially, the nanotubes inside the gel were isotropic and the sample under cross-polarizers appeared dark as shown in Fig. 6(a). The slow vacuuming of the chamber caused water from the center of the samples being extruded to the open ends of the capillary tubes and being evaporated off. The SWNTs-NIPA gel then started to shrink at the middle of the capillary tubes in width and thickness, as shown in Fig. 6(b). The flow out of water caused the nanotubes to align along the flow direction of water (the long axis of the capillary tubes) and the shrunk region became birefringent, shown in Fig. 6(b). Eventually most of the water was extruded from the gel and the entire gel became birefringent. Typical sample dimensions before and after water extrusion were (length \times width \times thickness) ~ 4 cm \times 4 mm \times 0.2 mm and ~ 2.8 cm \times 2 mm \times 0.1 mm, respectively. By varying the rate of water extrusion from the SWNTs-NIPA gels and/or the initial nanotube concentrations in gel, we were able to align SWNTs alone or to make small ropes of SWNTs as shown in Fig 6(c).

III. Method 3: Magnetic field induced alignment of nanotubes in NIPA gels To align nanotubes inside NIPA gels, we placed capillary tubes with SWNTs-NIPA gel inside a super-conducting magnet while the gel was polymerizing. The applied magnetic field aligned the nanotubes along the magnetic field before the nanotubes got locked into position by the NIPA gels. The entire sample looked strongly birefringent under cross-polarizers indicating high degree of alignment of nanotubes in the gels. Surprisingly, we also found that the nanotubes chained up to form "nanotube needles" under the magnetic field. In Fig. 7, we show such an image for a sample with initial SWNTs concentration of 0.78 mg/ml. The formation of nanotube needles or their density depended on the initial nanotube concentrations in gel, applied magnetic field strength (from 1 Tesla to 9 Tesla) and the gel viscosity. We changed the gel viscosity by varying the NIPA monomer and the cross-linker concentrations. Lower gel viscosity allowed the nanotubes to move in the gel during the polymerization process to form long needles. Shrinking this gel did not destroy the nanotube needles, rather increased their number density and also slightly increased the birefringence of the sample.

Other gels and suspending materials We have also suspended SWNTs in water, poly(methyl methacrylate) gel (PMMA) and poly(vinyl acetate) gel (PVA). In water we obtained ropes with a length distribution from 30 μ m to 2 cm. In PMMA and PVA gel, SWNTs formed similar structures as those formed in NIPA gels.

III. POTENTIAL APPLICATIONS

Our *nematic nanotube gels* can be use to create high quality composites for various applications. We list few examples below.

Polymer composites Currently we have created nanotube composites in NIPA gels, which is a semi-crystalline polymer. Nanotube can also be similarly dispersed and aligned in various other semicrystalline polymer gels such as polyvinyl acetate.(PVA), polymethyl methacrylate (PMMA) etc. Our alignment approach is very useful because we can just align the nanotubes or create aligned needles in polymeric gels. While aligned tube increase the strength and thermal properties of composites, composites with aligned nanotube needles can be very good heat dissipator.

Epoxies with nematic nanotube gels It is difficult to incorporate nanotubes in epoxies at high concentration. Our polymerized and shrunk nematic nanotube gels can ready be incorporated into epoxy used in semiconductor industries.

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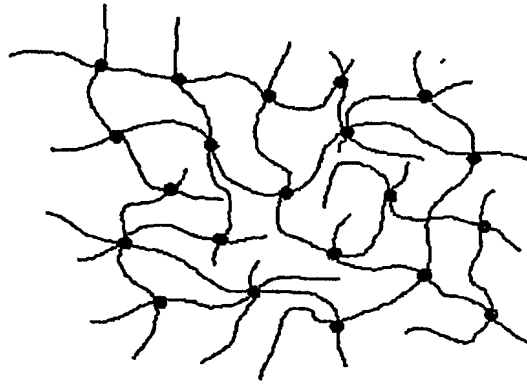


FIG. 1: A schematic of the NIPA gel structure after NIPA monomer polymerized in the presence of gel initiator and cross-linker at 295 K. At this polymerization temperature, we expected the gel to be homogeneous [28].

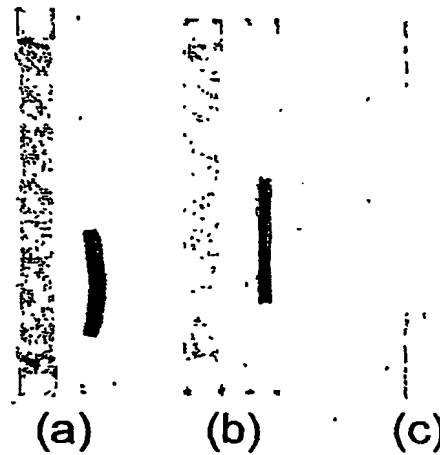


FIG. 2: Capillary tubes containing SWNTs-NIPA gels before and after shrinking. The volume change before/after was $\sim 8\times$. Capillary tubes containing initial nanotube concentration of 0.78 mg/ml (a) and 0.23 mg/ml (b) appeared dark because nanotubes absorb light. We also prepared a NIPA gel sample with no nanotube but just the surfactant (c) that were used to stabilize the nanotubes (NaDDBS) to compare the shrinking of NIPA gel with nanotubes. It is evident that NIPA gel shrank almost the same ratio with or without nanotubes in them.

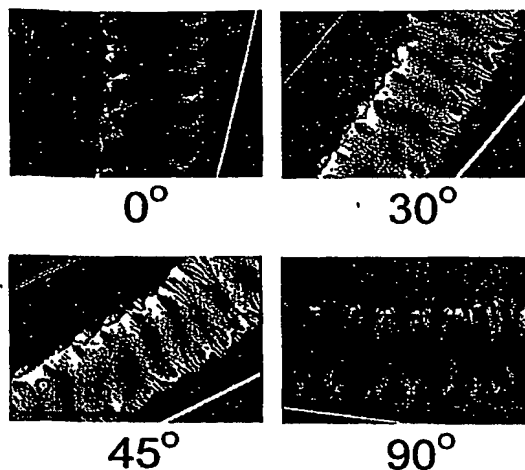


FIG. 3: Birefringence images of a sample with initial nanotube concentration of 0.78 mg/ml at different angles after it was allowed to sit for 4 days. Images were taken with a fixed microscope bulb intensity and video gain and offset. Maximum birefringence was found when the sample was oriented 45 degrees with respect to the input polarizer pass axis. Liquid crystal like defects were observed near the edges with the sample; clearly visible when the sample was in vertical (0 degree) or horizontal (90 degrees) orientations. Greater tube alignment was observed near the gel edges. Evidently the director tends to align near the walls, perhaps as a result of boundary effects. We believe that the central dark regions were disordered..

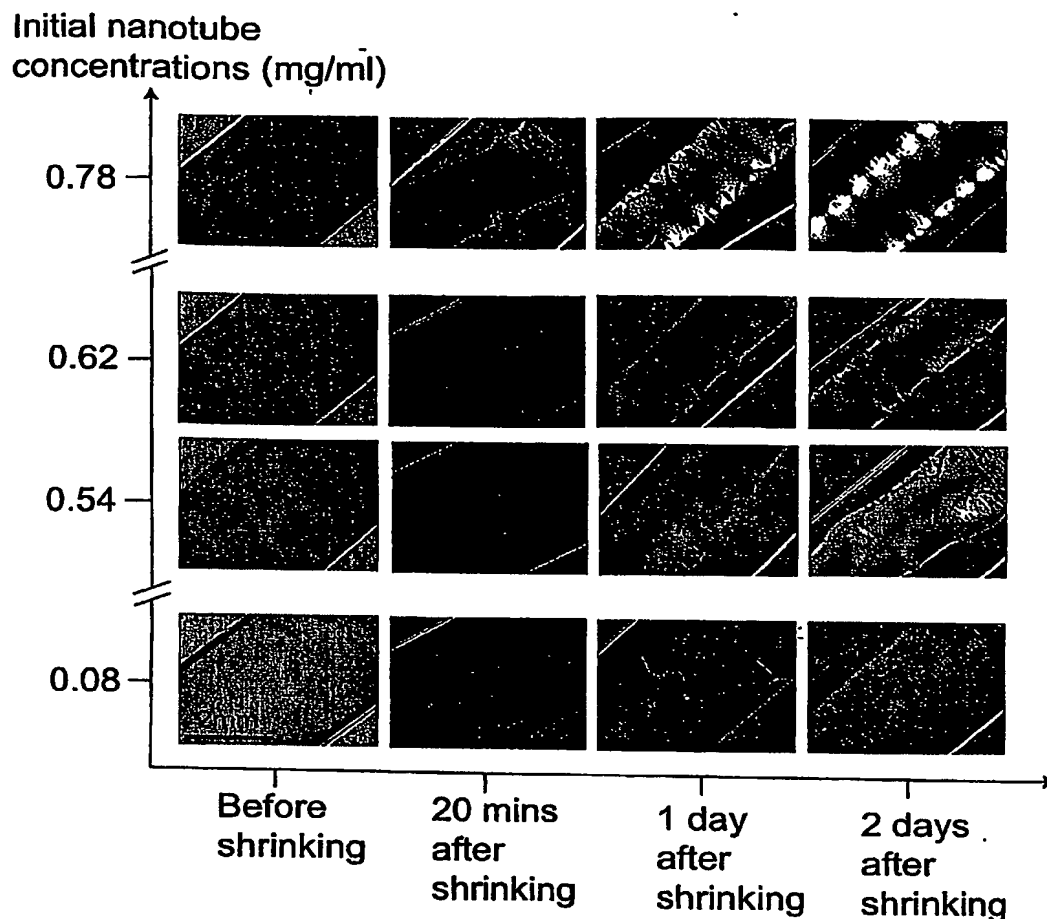


FIG. 4: A summary of the effects of time and nanotube concentration on the alignment of nanotubes in NIPA gels. The bulb intensity and video gain and offset were kept fixed. All of the samples were isotropic before shrinking. We only observed birefringence in samples after they were shrunk. Twenty minutes after shrinking only the highest initial concentration (0.78 mg/ml) sample exhibited birefringence. The nanotubes started to align in other samples slowly as time progressed. Alignment clearly started at the edges of the sample and migrated inward. After one day the sample critical concentration for birefringence near the edge was approximately 0.54 mg/ml. After 2 days the critical concentration for birefringence had dropped further.

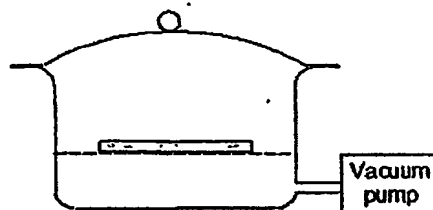


FIG. 5: Capillary tubes with SWNTs-NIPA gel were placed inside a vacuum jar and water from the sample was slowly extruded using a vacuum pump.

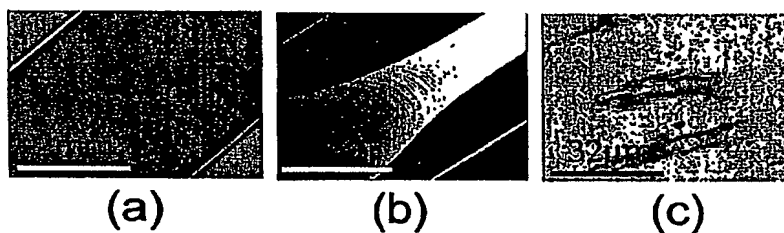


FIG. 6: The nanotubes inside NIPA gel were isotropic before water extrusion at 0.46 mg/ml (a). As water was extruded from the gel using a vacuum pump, nanotubes started to align along the flow direction of water and the sample became birefringent (b). At high enough initial concentration of nanotubes (0.46 mg/ml) in gel and after significant extrusion of water, some of the aligned nanotubes formed small ropes within the gel (c). The image (c) is a bright-field image at a higher magnification.



FIG. 7: The nanotubes aligned inside the NIPA gel in a 9 Tesla magnetic field. We also noticed that some of the tubes chained up and formed "nanotube needles".

Alignment of single-wall carbon nanotubes: Nematic nanotube gels

Abstract

We have developed three novel methods of aligning single-wall carbon nanotubes (SWNTs) inside a gel matrix; we call these materials *nematic nanotube gels*. To induce alignment of SWNTs in gels, SWNTs were dispersed at low concentration (≤ 0.78 mg/ml) in an aqueous N-isopropyl acrylamide (NIPA) gel precursor [1], and then polymerization was initiated by chemical means at 295K. The pre-gel solutions were then loaded into rectangular capillary tubes and allowed to polymerize at 295 K. The polymerization process completed in ~ 1 hour. In our first method of creating nematic nanotube gels, the gel volume was reduced substantially by increasing its temperature; this volume phase transition arises when the polymer network in the gel becomes hydrophobic and water is expelled (and removed). If the initial nanotube concentration was sufficiently large, then the tubes aligned locally. In our second method, we slowly evaporated water out of the SWNTs-NIPA gel through the open ends of the capillary tubes. The flow of water out of the capillary tubes caused the nanotubes to align along the flow direction of water (the long axis of the capillary tubes). In our last method, we placed the capillary tubes with SWNTs-NIPA gel inside a magnet immediately after the initiation of polymerization for the duration of the polymerization process. The nanotubes get aligned by the magnet and get locked in place by the gel. In the last approach, we could just align the nanotubes, make needles with multiple tubes or long aligned ropes by varying the magnetic field strength, gel viscosity and polymerization time.

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